## Further cubic spin-orbit terms

The following discussion was prepared with major input from Hans-Andreas Engel, and we are grateful for his contribution.

In 3D zincblende crystals, the lowest order in k spinorbit coupling term for conduction band electrons consistent with the crystal symmetries is

$$\mathcal{H}_{\text{Dresselhaus}} = \gamma(\sigma_x k_x (k_y^2 - k_z^2) + \text{c.p.})$$
(1)

where c.p. indicates cyclic permutations and x, y, z are the cubic crystalline directions [S1]. When electrons are confined on a (001) plane,  $k_z^2$  can be sent to  $\langle k_z^2 \rangle$  and the 3D Dresselhaus term produces the 2D linear and cubic Dresselhaus terms,

$$\mathcal{H}_{001} = \gamma \langle k_z^2 \rangle (\sigma_y k_y - \sigma_x k_x) + \gamma (\sigma_x k_x k_y^2 - \sigma_y k_y k_x^2).$$
(2)

Asymmetric confinement in the (001) plane also produces spin-orbit coupling of the Rashba type [S2],

$$\mathcal{H}_{\text{Rashba}} = \alpha (k_y \sigma_x - k_x \sigma_y). \tag{3}$$

These three terms together give the effective Hamiltonian for 2D conduction electrons in Eq. 1 of the main text.

In a 2D zincblende system on a (001) surface, there are further cubic spin-orbit terms originating from both bulk and structure inversion asymmetries [S3]. The extra contribution from bulk inversion asymmetry is  $\gamma_1(k_x^3\sigma_x - k_y^3\sigma_y)$  while the cubic Rashba terms are  $\alpha_1(k_x^3\sigma_y - k_y^3\sigma_x) + \alpha_2(k_yk_x^2\sigma_x - k_xk_y^2\sigma_y)$ .

Winkler showed that  $\gamma_1$  (which he calls  $b_{53}^{6c6c}$ ) is much less than  $\gamma$  [S4]. Specifically, using his formulae and parameters for GaAs gives  $\gamma/\gamma_1 \approx 10^3$  for  $k_F = 0.19 \text{ nm}^{-1}$ . In a similar calculation, von Allmen considered an axial cubic Dresselhaus term of the form  $\gamma_2 k^2 (k_x \sigma_x - k_y \sigma_y)$ and showed that  $\gamma/\gamma_2 \approx 20$ -400, depending on the model used [S5].

We are unaware of estimates of the magnitudes of  $\alpha_1$ and  $\alpha_2$  in GaAs. Yang and Chang recently showed that in a 15 nm-wide GaAs/AlGaAs quantum well, the nonlinear Rashba terms do not become significant until considerably higher electron densities than considered in this paper [S6].

An approximation of the cubic Rashba terms can also be given using the  $8 \times 8$  Kane model, which considers the *s*-type conduction band and the *p*-type valence bands. The model is parameterized by the band gap  $E_0$ , the energy of the split-off holes  $\Delta_0$ , and the matrix element *P* of the momentum (times  $\hbar/m_0$ , where  $m_0$  is the free electron mass) between the *s*- and *p*-type states. When an external electric potential is applied across the sample, the conduction band Hamiltonian contains a term

$$\mathcal{H}_{\text{ext}} = \lambda \,\boldsymbol{\sigma} \cdot (\mathbf{k} \times \nabla V) \,. \tag{4}$$

For a system confined along the z-direction, one can take the expectation value  $\langle \mathcal{H}_{\text{ext}} \rangle$  along that direction. Noting that the only contribution of the confinement field is  $\propto \langle \nabla_z V \rangle$  and for  $\lambda$  a constant, one obtains the k-linear Rashba Hamiltonian  $\mathcal{H}_{\text{Rashba}}$ . If we consider higher order corrections to  $\lambda$ , we can write  $\lambda = \lambda^{(0)} + \lambda^{(1)}k^2$ . The  $\lambda^{(1)}$ term gives a term  $\mathcal{H}_{\text{Rashba},c} = \alpha_3 k^2 (k_y \sigma_x - k_x \sigma_y)$ , which gives the axial  $(C_{\infty v})$  approximation to the cubic Rashba terms.

Using third-order perturbation theory [S4, S7],

$$\lambda^{(0)} = \frac{P^2}{3} \left[ \frac{1}{E_0^2} - \frac{1}{(E_0 + \Delta_0)^2} \right].$$
 (5)

Fifth-order perturbation theory gives [S8]

$$\lambda^{(1)} = -\frac{P^4 \Delta_0 \left(24E_0^3 + 41E_0^2 \Delta_0 + 26E_0 \Delta_0^2 + 6\Delta_0^3\right)}{9E_0^4 \left(E_0 + \Delta_0\right)^4}.$$
(6)

In GaAs, these yield  $\lambda^{(0)} = 5.3$  Å<sup>2</sup> and  $\lambda^{(1)} = -870$  Å<sup>4</sup>.

Because this estimate does not include edge effects, it only provides a rough estimate for the coupling constants  $\alpha$ ,  $\alpha_3$ . We can, however, use Eqs. 5 and 6 to estimate the ratio of the strengths of the linear and cubic Rashba terms. For  $k = 0.19 \text{ nm}^{-1}$  corresponding to the sample considered in the main text,

$$\frac{k^2 \lambda^{(1)}}{\lambda^{(0)}} \approx \frac{1}{17}.$$
 (7)

We are interested, however, in the relative sizes of the cubic Dresselhaus and the cubic Rashba. In the sample considered, the linear Dresselhaus and the linear Rashba strengths appear to be almost the same [S9], and we can estimate that the linear Dresselhaus is about twice the strength of the cubic Dresselhaus (i.e.,  $\langle k_z^2 \rangle / k_F^2 \approx 2$ , using the results in Ref. [S9]), giving  $\gamma/\alpha_3 \approx 8$ .

It is possible that a more careful calculation of the cubic Rashba strengths, including band offsets and the full crystal symmetry, would modify these results substantially, but these results indicate that the cubic Dresselhaus term considered in the main text is the dominant cubic contribution to the spin-orbit coupling of GaAs quantum dots.

Regardless of their strengths, all of these cubic terms contribute to  $a_{\parallel}$  similarly to the cubic Dresselhaus term discussed in the main text. We neglect the other cubic contributions, but their inclusion would only increase the constraint on  $\gamma$ .

## Refitting var g data

Fig. 1 shows the Zumbühl et~al.vargversus  $B_{\parallel}$  data discussed in the main text [S31]. Also shown are the original curve fit and our refit with  $a_{\parallel}$  constrained to be greater than 8.1. This constrained fit clearly does not match the data. We thank Dominik Zumbühl for making the data available to us.



FIG. 1: (color) Variance in conductance as a function of inplane field for quantum dot of area  $A = 8 \ \mu m^2$  and electron density  $n = 5.8 \times 10^{15} \ m^{-2}$ , from Ref. [S31]. A small per-pendicular magnetic field is applied to break time reversal symmetry. The original fit to the data is shown along with our refit constraining only  $a_{\parallel} \geq 8.1$  and  $\tau_{\phi} = 0.39$  ns (the value found from  $\langle g \rangle$ ).

## Value of $\gamma$ in GaAs

We include here a table of values of  $\gamma$  from both experiment and theory.

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otherv	wise specified.	). An results are in burk GaAs unless
Ref	$ \gamma $	Technique/Comments
		EXPERIMENT
[S10]	24.5	Optical orientation using Dyakonov-Perel (DP) spin relaxation time. Conduction electrons in p-type GaAs.
[S11]	20.9	Same as [S10].
[S12]	26.1	GaAs/AlGaAs heterostructure magneto- conductance measurements. Sets $k_z^2 \to 0$ instead of $\langle k_z^2 \rangle.$
[S13]	23.5	Raman scattering from 180Å GaAs quantum well (QW). Does not include Rashba term.
[S14]	34.5	Same as [S13]. No Rashba term.
[S15]	$16.5 \pm 3$	Raman scattering from GaAs/AlGaAs heterostructure.
[S16]	11.0	Raman scattering from asymmetric GaAs/AlGaAs QW. More data than earlier.
[S9]	$28 \pm 4$ ; $31 \pm 3$	${\rm GaAs/AlGaAs}$ heterostructure magneto-conductance measurements.
		THEORY
[S17]	9;8.5	$14 \times 14 \mathbf{k} \cdot \mathbf{p}$ theory; Linear Muffin Tin Orbitals (LMTO). Room temperature band gap.
[S18]	19:30	$8 \times 8 : 14 \times 14 \mathbf{k} \cdot \mathbf{p}$ models.
[S19]	28	$14 \times 14 \mathbf{k} \cdot \mathbf{p}$ .
[S20]	27.57; 21.29	$14 \times 14$ <b>k</b> · <b>p</b> . Bulk GaAs ; Ga. <sub>65</sub> Al <sub>.35</sub> As/GaAs heterostructure.
[S21]	14.0	$14 \times 14 \ \mathbf{k} \cdot \mathbf{p}.$
[S22]	14.9; 28.2	LMTO ; $16 \times 16 \mathbf{k} \cdot \mathbf{p}$ .
[S23]	24.12	$14 \times 14 \ \mathbf{k} \cdot \mathbf{p}.$
[S5]	18.3 ; 7.6 ; 36.3	$14\times14~{\bf k}\cdot{\bf p}$ in bulk GaAs; bulk Al_{0.35}Ga_{0.65}As ; GaAs/AlGaAs superlattice.
[S24]	19.8	$\mathbf{k} \cdot \mathbf{p}$ .
[S25]	8.9	$sp_3s^*$ tight binding (TB) model of a GaAs/AlAs superlattice.
[S26]	8.5	TB model of 100 Å GaAs/AlAs QW.
[S27]	24.21	$14 \times 14 \ \mathbf{k} \cdot \mathbf{p}.$
[S28]	10; 27.5	$sp_3s^*$ TB model ; $\mathbf{k} \cdot \mathbf{p}$ .
[S4]	27.48	$14 \times 14 \mathbf{k} \cdot \mathbf{p}$ theory. Notes that value is reduced to 19.6 in higher order perturbation theory (p. 74).
[S29]	23.6	TB model used to refine $14 \times 14 \mathbf{k} \cdot \mathbf{p}$ .
[S30]	8.5	Quasiparticle self-consistent GW method (QPscGW).

TABLE I: Magnitude of Dresselhaus spin-orbit coupling const ot =

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